Responses to the Anonymous Referee 1 comments

Referee #1 Evaluations

The work by Haque et al presents a range of organic compounds in atmospheric aerosols from a year-long campaign in South Korea. The authors used well established analytical techniques for their analysis. The experimental procedure is well designed and in general the data is well presented. My detailed comments are listed below.

Response: Thanks for the careful reading and valuable suggestions to improve the scientific content of the manuscript. Following the reviewer's comment, we have carefully made corrections in the revised MS. Our responses to all comments made by the reviewer are given below. Please refer to the revised MS, in which changes are highlighted in yellow.

Comment: First of all, although the work is of relatively good quality (as can be judged from the draft); I felt it is largely of a routine nature (at least the way it is presented). I believe that such type of publications can be useful but if considered for publication at ACP they need to go under the Measurement report's category.

Response: We respectfully do not comply with the reviewer's argument but would like to highlight the two key features that make this study suitable for publication in ACP under the Research article category.

1. Here, we present a new finding on the contribution of coal combustion sources in East Asia in controlling the atmospheric levels of levoglucosan (*Lev*) apart from the traditional biomass/biofuel burning emissions. This is based on the prevailing linear relationship between the radiocarbon based nonfossil-EC and *Lev* in the year-round TSP samples collected from the Korean Climate Observatory at Gosan (KCOG) site in Jeju Island. The Gosan supersite is the best location to understand how the chemical composition of source-emissions from East Asia affects the outflow regions in winter and spring. Recent studies have highlighted the potential contribution of *Lev* from residential coal combustion in China (Yan et al., 2018), with estimated annual emission of ~2.2 Gg of *Lev* from domestic coal combustion (Wu et al., 2021). Given this background information, the prevailing significant linear relationship between *Lev* and nonfossil-EC (P-value < 0.05) over the KCOG clearly emphasizes the need for reconsideration of the previous assessments on the impact of biomass burning in East Asian outflow to the western North Pacific.

2. Another important highlight is the detailed study on the atmospheric abundances of biogenic SOA tracers along with those of other primary biological particles from the KCOG for the year 2013. This dataset is further compared with the molecular distributions and relative abundances of these organic tracers in the TSP samples collected over Gosan during 2001, a decade ago (Fu et al., 2012). This comparison allows us to better understand the regional changes in the emission sources (*e.g.*, fugitive dust, biomass burning, and fossil-fuel combustion) on a decadal basis.

These points have been added in the revised MS. Please see lines 682-697.

Major comment:

Comment: I would like to know if the authors considered atmospheric stability of the measured organic compounds especially in the light of a long-range transport or/and correlation and statistical analysis of the reported species. For example, it has been demonstrated that levoglucosan can decay in particles exposed to OH. ''*The extent of decay ranged from 20 to 90% and was strongly correlated to the integrated OH exposure. Increased relative humidity did not enhance or impede reaction. Relative kinetics indicate that levoglucosan has an atmospheric lifetime of 0.7–2.2 days when biomass burning particles are exposed to 1 × 106 molecules* cm^{-3} *of OH (typical average summertime conditions). This implies that levoglucosan reacts with OH on a timescale similar to that of transport and deposition, which has important implications for the use of levoglucosan as a tracer for biomass burning emissions in source apportionment studies'' Hennigan et al. 2010.*

I trust the above also applies to other measured organic compounds for some of which the information on reactivity is not available. My question to the authors - how does this affect the presented results? The authors present correlation of different organic compounds and discuss their potential similar source based on these correlations and statistical analysis. Does the stability of considered organic compounds can play a role in the observed absence of correlation? The same applies to the discussion on long range transport of the observed organics. I doubt that stability of considered sugars in Figure 4 is the same for all compounds.

Response: As the reviewer rightly pointed out that levoglucosan (*Lev*) undergoes a photooxidation with OH radicals during atmospheric transport (half-life: <2.2 days) and, this would cause lower abundances of this anhydrosugar (Hennigan et al., 2010). *Lev* concentration measured over the KCOG includes contributions from both fossil and nonfossil

sources in the East Asian outflow. In such a case, we found the moderately significant correlations of *Lev* with nonfossil EC (i.e., biomass burning) and fossil-EC (e.g., Miocene lignite coals) over the KCOG. It is very likely that this weakened relationship is because of the photo-degradation of *Lev* during atmospheric transport. This result would be an otherwise high atmospheric abundance of *Lev* and its pronounced linear relationships with the nonfossil- and fossil-EC, which implies a much stronger impact of both these source emissions in the East Asian continental outflow in winter and spring. This is also applicable for low concentrations and poor correlations of other organics such as primary saccharides and BSOA tracers. Considering the feasibility of photooxidation, it is likely that secondary OA undergoes much faster cycling than the primary sugar compounds. This would mean a slight underestimation of BSOA over the KCOG in the East Asian outflow and their atmospheric abundances over Gosan reflect a lower limit.

These explanations have been added in the revised MS. Please see lines 370-374, 452-453, 458-460, and 676-681.

Comment: Moreover, besides anhydrosugars, the authors state that ''*Cis-pinonic acid showed poor correlation with pinic acid (r = 0.35, p = 0.12) (Figure 6f), suggesting that they formed from different monoterpenes such as α-pinene, β-pinene, or δ-limonene*.'' Such assumptions can be made only if the measurements are taken very close to their emission sources **and/or** assuming that both compounds have similar atmospheric stability or reactivity. Is it the case for all considered compounds in this study? I believe this needs to be considered when discussing correlations or long range transport.

Response: We comply with the reviewer's view. Likely, the extent of loss of BSOA by the photooxidation during transport would also cause such poor correlations, influencing their abundances over KCOG. We have added a few sentences in the revised MS. Please see lines 476-478 and 499-501.

Minor comment:

Comment: It is hard to read Figure 2 (especially Figure 2a). Please either split the concentration axis so that all minor compounds can be clearly seen in the plot or move it into the SI and increase the size of the figure.

Response: Following the reviewer's suggestion, we have added an additional figure in the supplement for the visualization of minor compounds in the temporal plot. Please see Figure S2.

Reference:

Hennigan, C.J., Sullivan, A. P., Collett, J. L., Robinson, A. L.: Levoglucosan stability in biomass burning particles exposed to hydroxyl radicals, Geophys. Res. Lett., 37, https://doi.org/10.1029/2010GL043088, 2010.

Response: Added in the reference list of the main text.